Modeling of Plutonium (Pu) Production in Foreign Nuclear Fuel Cycles (FNFC)

MTV Workshop, 2020

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Introduction and Motivation

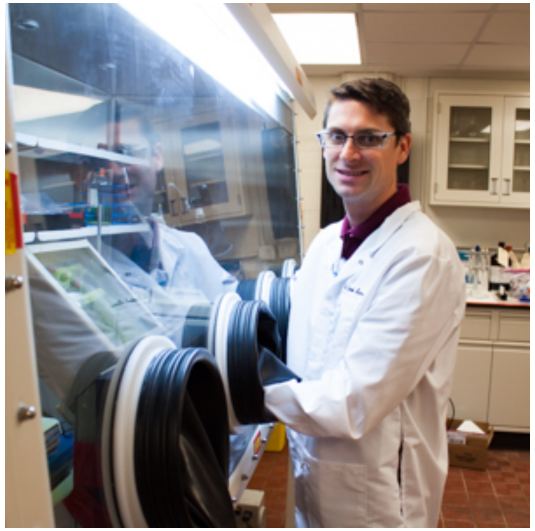
- Nuclear weapon states outside of the NPT and facilities not under the IAEA safeguards
  - Nuclear security (non-state actors) and safeguards (state actors) concerns of plutonium production in these states

- Motivation: Identify forensics signatures and proliferation identifiers for these Nuclear Fuel Cycles

Mission Relevance

- Develop an enhanced and innovative FNFC monitoring method
  - Nuclear forensics signatures and proliferation identifiers of FNFC
    - Modeling and experimental efforts of Pu and ^{233}U production reactors and associated fuel cycle facilities
    - Accurate methods for discriminating source of Pu and ^{233}U (for nuclear security and safeguards applications)

Technical Work Plan in Collaboration with National Labs



- Task 1: High-fidelity computational modeling of a set of FNFCs
  - U and Th fueled reactors (research and power) with fuel reprocessing (aqueous and electrochemical)
- Task 2: Experiments to point-validate the results from the modeling efforts.
  - Low-fluence neutron irradiation of U and Th surrogates using neutron spectrum modifying capsules to produce milligram quantities of Pu and ^{233}U
  - Radiochemical separations and contaminant analyses using alpha, gamma and mass spectrometry
- Task 3: A maximum likelihood data analytics method to discriminate source reactor-type of Pu and ^{233}U
  - Fuel burnup and time since irradiation predictions

$$L(M|r_{mes}) \propto f(r_{mes}|M) = \prod_{j=1}^{n} \frac{1}{\sigma_{j,sim}\sqrt{2\pi}} exp\left\{-\frac{(r_{j,mes}-r_{j,sim})^2}{2\sigma_{j,sim}^2}\right\}$$

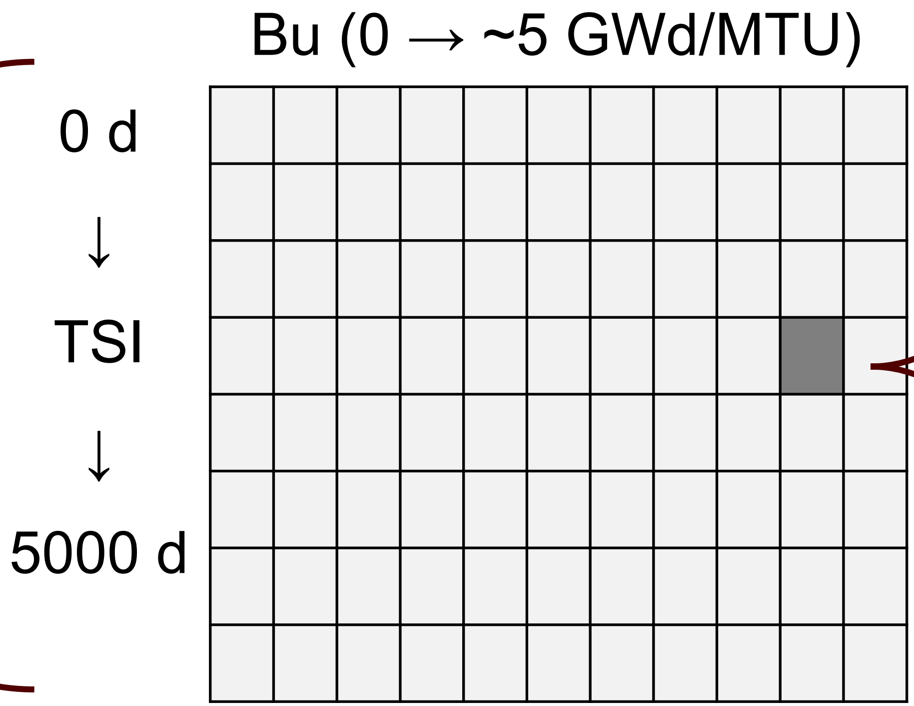
$$Log\ L(M|r_{mes}) = \sum_{j=1}^{n}\left[log\left(\frac{1}{\sigma_{j,sim}\sqrt{2\pi}}\right) - \frac{(r_{j,mes}-r_{j,sim})^2}{2\sigma_{j,sim}^2}\right]$$

$$\sigma_{Log\ L}^2 \cong \sum_{j=1}^{n}\left(\frac{(r_{j,mes}-r_{j,sim})}{\sigma_{j,sim}^2}\right)^2 \times \left(\sigma_{j,mes}^2 + \sigma_{j,sim}^2\right)$$

Task1: Simulated Reactor Library

Reactor Type
PWR(2.35%)
PWR(3.4%)
PWR(4.45%)
FBR
PHWR
NRX
MAGNOX
HFIR
MURR

Reactor Model Burnup and Time Since Irradiation Ratio Set Matrix



Ratio Set (Bu, TSI)
^{137}Cs / ^{133}Cs
^{134}Cs / ^{137}Cs
^{135}Cs / ^{137}Cs
^{136}Ba / ^{138}Ba
^{154}Eu / ^{153}Eu
^{150}Sm / ^{149}Sm
^{152}Sm / ^{149}Sm
^{240}Pu / ^{239}Pu
^{241}Pu / ^{239}Pu
^{242}Pu / ^{239}Pu

$$Log\ L(M, Bu, TSI | r_{mes}) = \sum_{j=1}^{n} \left[log\left(\frac{1}{\sigma_{j,sim}\sqrt{2\pi}}\right) - \frac{\left(r_{j,mes} - r_{j,sim}\right)^2}{2\sigma_{j,sim}^2} \right]$$

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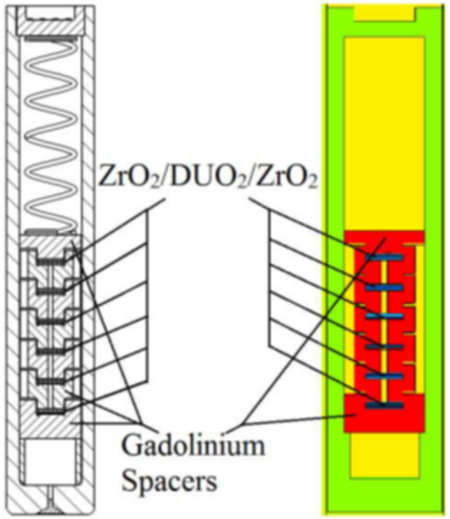
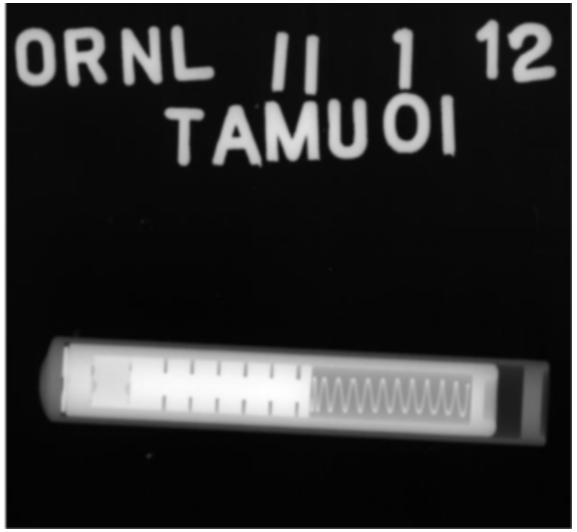
Task 2: Experimental Irradiation at HFIR

- Depleted UO_2 fuel samples irradiated at HFIR
- Gadolinium irradiation capsule
- Burnup = 4.36 ± 0.28 GWd/MTU
- Each pellet: 11 mg uranium, produced 200 μg plutonium with 87% ^{239}Pu
- TSI = 1601 days





HFIR irradiated material mass spectroscopy measured set of intra-element ratio values

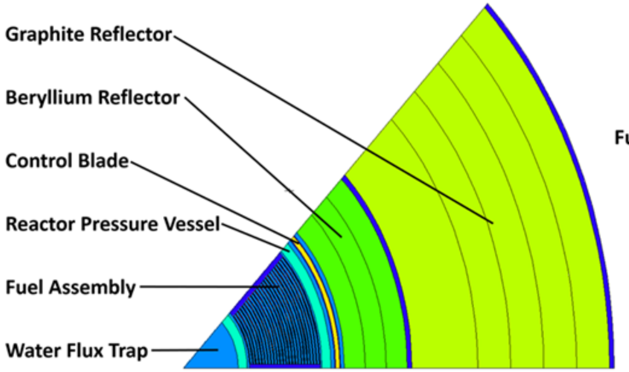
Ratio	Measured Value	Measurement Error
$^{137}Cs/^{133}Cs$	1.30×10^{0}	6.7%
$^{134}Cs/^{137}Cs$	3.74×10^{-3}	4.2%
$^{135}Cs/^{137}Cs$	4.25×10^{-1}	10%
$^{154}Eu/^{153}Eu$	4.67×10^{-2}	4.5%
$^{150}Sm/^{149}Sm$	3.23×10^{0}	2.7%
$^{152}Sm/^{149}Sm$	2.93×10^{0}	1.3%
$^{240}Pu/^{239}Pu$	8.28×10^{-2}	0.59%
$^{241}Pu/^{239}Pu$	3.30×10^{-2}	0.88%
$^{242}Pu/^{239}Pu$	1.88×10^{-3}	0.88%

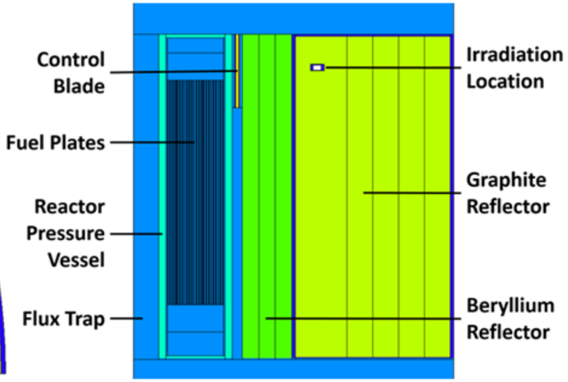
Task 2: Experimental Irradiation at MURR

- Natural UO_2 fuel samples irradiated at MURR
- Complex irradiation history
- Irradiation history and axial/radial location of samples known
- Burnup = 0.97 ± 0.03 GWd/MTU
- TSI = 318 days

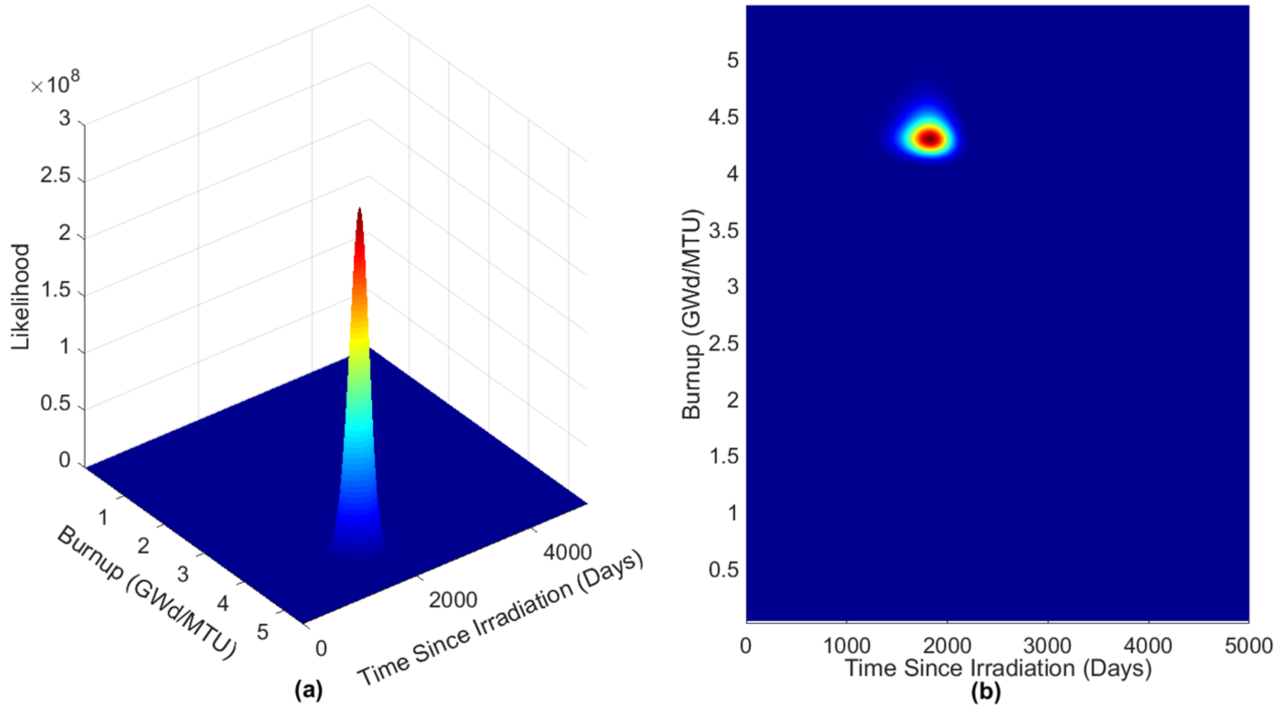
MURR irradiated material mass spectroscopy measured set of intra-element ratio values

Ratio	Measured Value	Measurement Error
$^{137}Cs/^{133}Cs$	9.75×10^{-1}	6.6%
$^{134}Cs/^{137}Cs$	3.84×10^{-3}	7.0%
$^{135}Cs/^{137}Cs$	2.95×10^{-1}	6.8%
$^{150}Sm/^{149}Sm$	9.88×10^{0}	6.7%
$^{152}Sm/^{149}Sm$	6.65×10^{0}	5.7%
$^{240}Pu/^{239}Pu$	4.77×10^{-2}	5.7%
$^{241}Pu/^{239}Pu$	2.40×10^{-3}	5.8%
$^{242}Pu/^{239}Pu$	5.99×10^{-5}	8.3%





Maximum Likelihood Results – HFIR Cont.



Results of Maximum Likelihood Analysis for the HFIR Irradiated Material (a) 3-D Likelihood Surface Map and (b) 2-D Contour Map for the Most Likely Reactor (HFIR)

Ratio	Measured Value	Selected HFIR Simulation Value	Selected /Mes
$^{137}Cs/^{133}Cs$	1.30×10^{0}	9.74×10^{-1}	0.73
$^{134}Cs/^{137}Cs$	3.74×10^{-3}	3.71×10^{-3}	0.99
$^{135}Cs/^{137}Cs$	4.25×10^{-1}	4.95×10^{-1}	1.16
$^{154}Eu/^{153}Eu$	4.67×10^{-2}	4.66×10^{-2}	1.00
$^{150}Sm/^{149}Sm$	3.23×10^{0}	3.33×10^{0}	1.03
$^{152}Sm/^{149}Sm$	2.93×10^{0}	2.46×10^{0}	0.84
$^{240}Pu/^{239}Pu$	8.28×10^{-2}	8.75×10^{-2}	1.06
$^{241}Pu/^{239}Pu$	3.30×10^{-2}	4.22×10^{-2}	1.28
$^{242}Pu/^{239}Pu$	1.88×10^{-3}	2.72×10^{-3}	1.44

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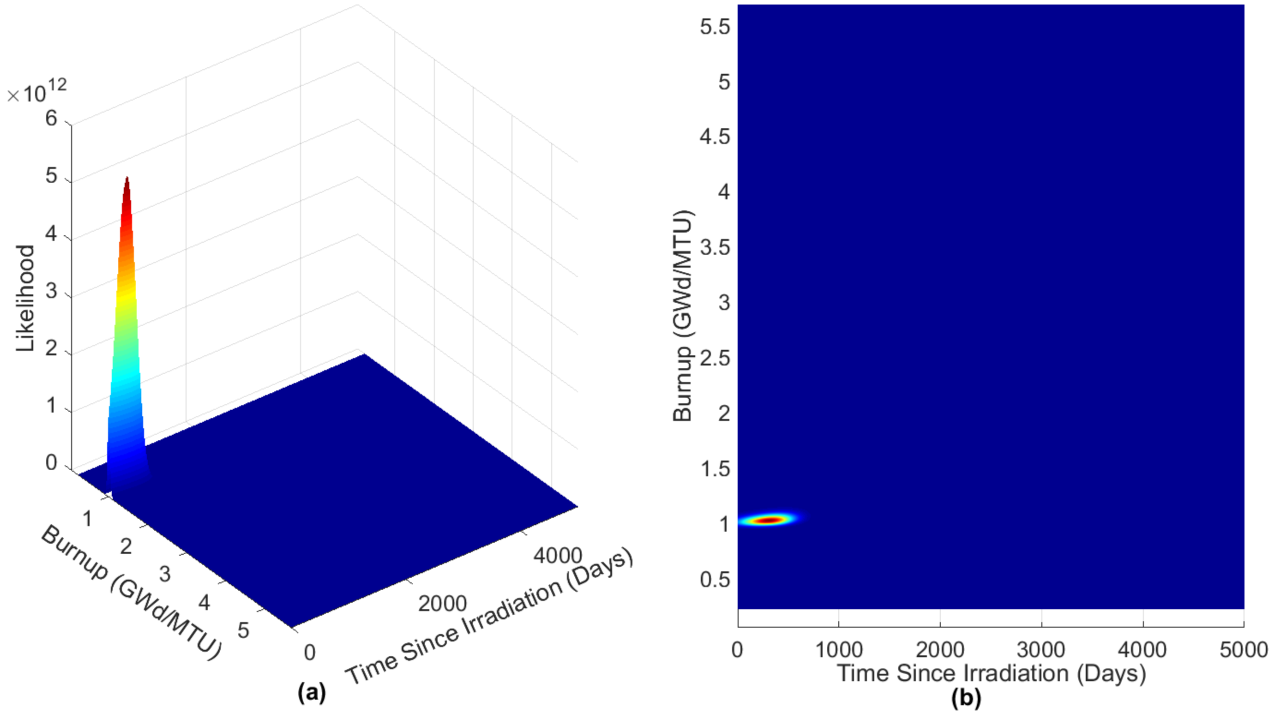
Maximum Likelihood Results – MURR Cont.



Results of Maximum Likelihood Analysis for the MURR Irradiated Material (a) 3-D Likelihood Surface Map and (b) 2-D Contour Map for the Most Likely Reactor (MURR)

Ratio	Measured Value	Selected MURR Simulation Value	Selected /Mes
$^{137}Cs/^{133}Cs$	9.75×10^{-1}	9.43×10^{-1}	0.97
$^{134}Cs/^{137}Cs$	3.84×10^{-3}	3.77×10^{-3}	0.98
$^{135}Cs/^{137}Cs$	2.95×10^{-1}	2.77×10^{-1}	0.94
$^{150}Sm/^{149}Sm$	9.88×10^{0}	1.02×10^{1}	1.03
$^{152}Sm/^{149}Sm$	6.65×10^{0}	6.20×10^{0}	0.93
$^{240}Pu/^{239}Pu$	4.77×10^{-2}	4.42×10^{-2}	0.93
$^{241}Pu/^{239}Pu$	2.40×10^{-3}	2.30×10^{-3}	0.96
$^{242}Pu/^{239}Pu$	5.99×10^{-5}	6.01×10^{-5}	1.00

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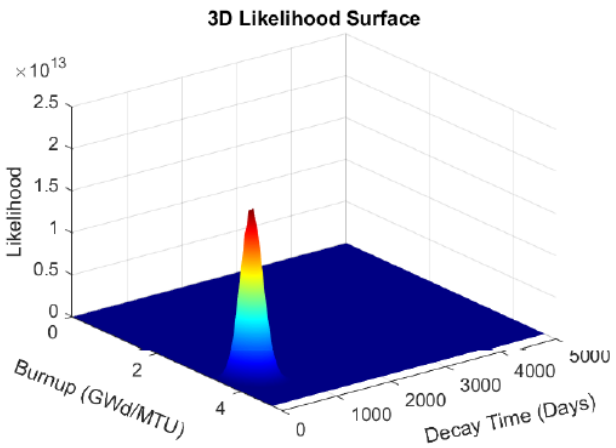
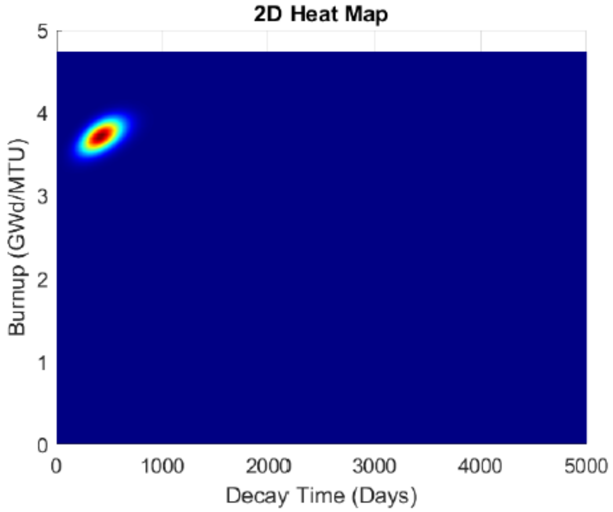
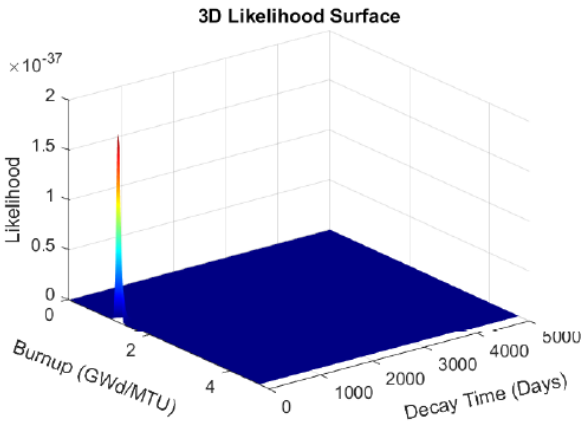
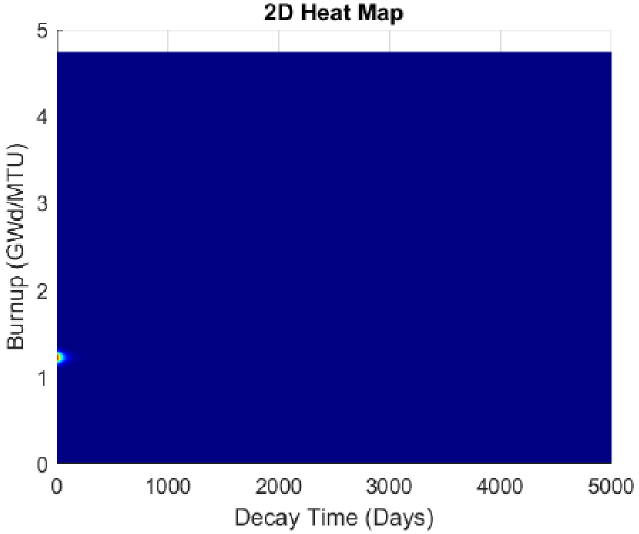




	Likelihood	Burnup (GWD/MTU)	Time (days)
PWR	2.07×10^{13}	3.74	429
PHWR	3.44×10^{-1}	1.28	1
FBR	0		

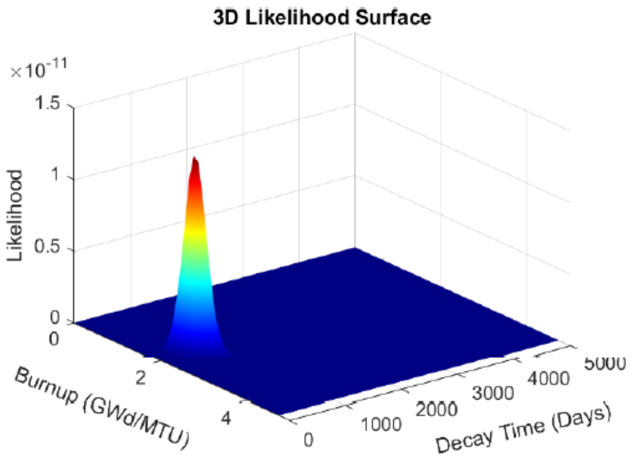
Spoof 1: 50% PWR (4 GWD/MTU) and 50% PHWR (1 GWD/MTU) with one year cooling period

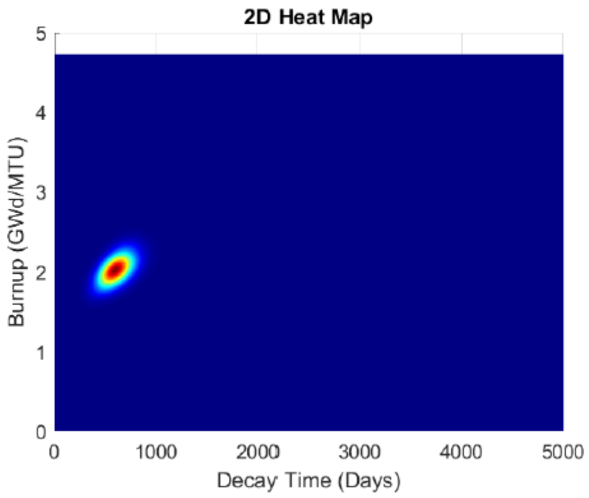




	Likelihood	Burnup (GWD/MTU)	Time (days)
PWR	1.88×10^{-37}	1.2	1
PHWR	1.44×10^{-67}	0.35	1
FBR	4.14×10^{-69}	0.92	1

Spoof 2: 50% PWR (4 GWD/MTU) and 50% FBR (2 GWD/MTU) with one year cooling period



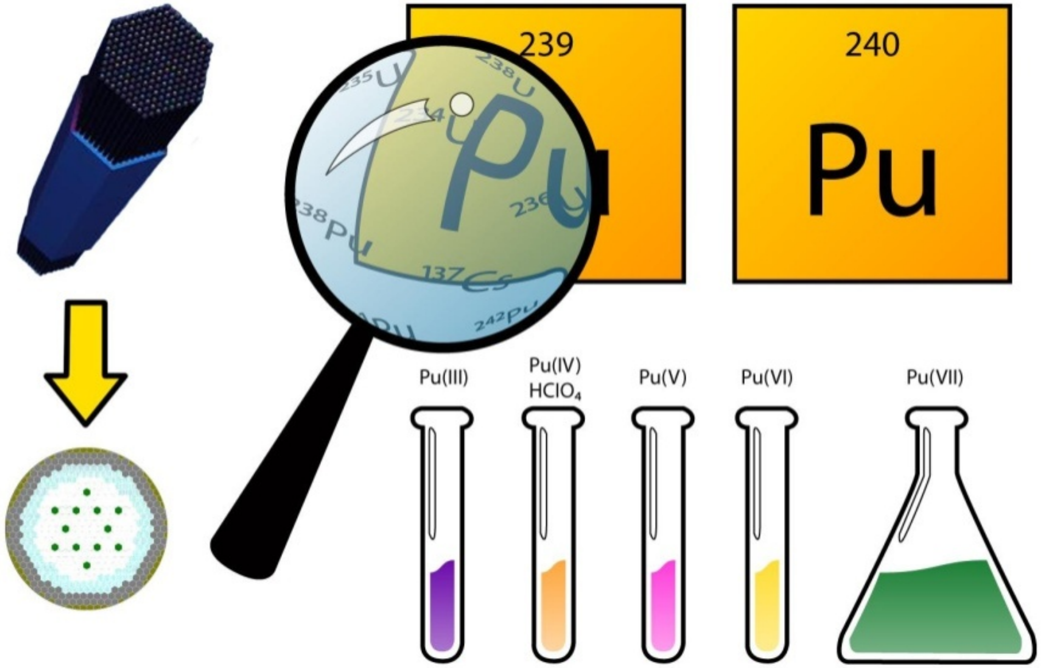


	Likelihood	Burnup (GWD/MTU)	Time (days)
PWR	9.44×10^{-75}	0.54	1
PHWR	4.60×10^{-92}	0.14	1
FBR	1.38×10^{-11}	2.01	592

Spoof 3: 50% PHWR (1 GWD/MTU) and 50% FBR (2 GWD/MTU) with one year cooling period

Expected Impact

- Successful completion of this project will enhance the US capability to monitor FNFCs
  - Application in environmental and wide area environmental sample analyses (Safeguards monitoring)
  - Application in nuclear security (Material out of regulatory control)
- A library for various combinations of FNFC facility operations that could produce Pu and ^{233}U



MTV Impact

- Two PhD students and one MS student
- Internships in LLNL and ANL and potential transitions as employees
- Faculty time in LLNL and ANL
- Workshops:
  - Reactor core physics simulation with MCNP6 for users with single user license
  - Radiochemistry analytical techniques
- Technology transitions
  - National lab collaborations and potential transition of the methodology
  - Collaborated with DHS in the past on similar project

Conclusion

- This project will enhance US capability to monitor FNFC
- We will produce a library for various combinations of FNFC facility operations that could produce Pu and ^{233}U. The data will be used to inform and validate specific facility models
- Nuclear engineering students with nuclear reactor core and fuel cycle modeling, simulation and radiochemical expertise for potential transition to national laboratories
- Workshops: To enhance MTV students/researcher expertise

Next Steps

- New experimental irradiation of LEU in MURR and analysis
- Fuel separation modeling in collaboration with ANL and LLNL
- Thorium irradiation modeling, separation and analysis

Acknowledgements

The Consortium for Monitoring, Technology, and Verification would like to thank the NNSA and DOE for the continued support of these research activities.

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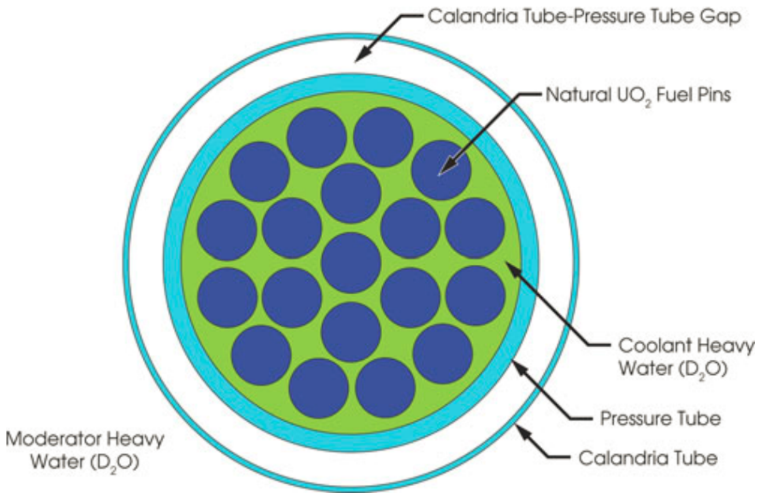
Backup Slides

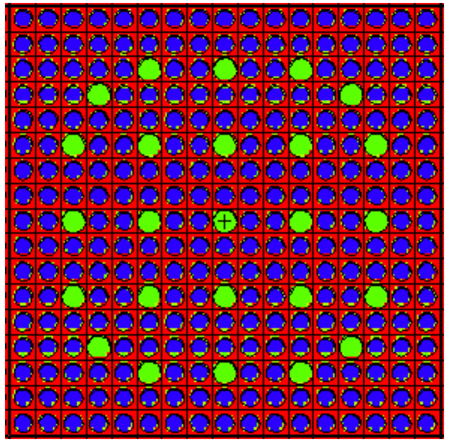
Reactor Library Model Characteristics

Reactor Model	Power (MWth)	Fuel Type (at.% ^{235}U)	Moderator	Coolant
PWR (2.35%)	3400	UO_2 (2.35)	Light Water	Light Water
PWR (3.4%)	3400	UO_2 (3.4)	Light Water	Light Water
PWR (4.45%)	3400	UO_2 (4.45)	Light Water	Light Water
FBR (blanket)	1250	UO_2 (0.25)	-	Liquid Sodium
PHWR	756	UO_2 (0.72)	Heavy Water	Heavy Water
NRX	40	UO_2 (0.72)	Heavy Water	Heavy Water
MAGNOX	25	U metal w/ 0.5% Al (0.72)	Graphite	Carbon Dioxide
HFIR (irradiation)	85	UO_2 (0.25)	Light Water	Light Water
MURR (irradiation)	10	UO_2 (0.72)	Light Water	Light Water

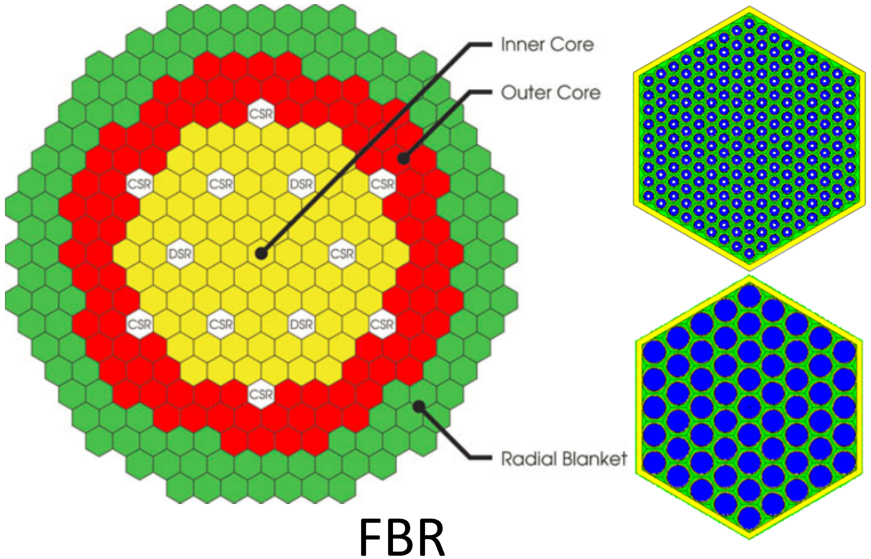
MCNP Reactor Models



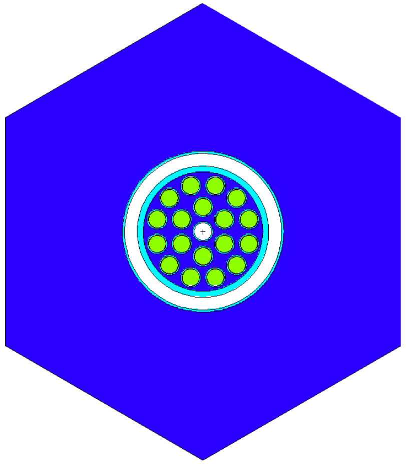
PHWR



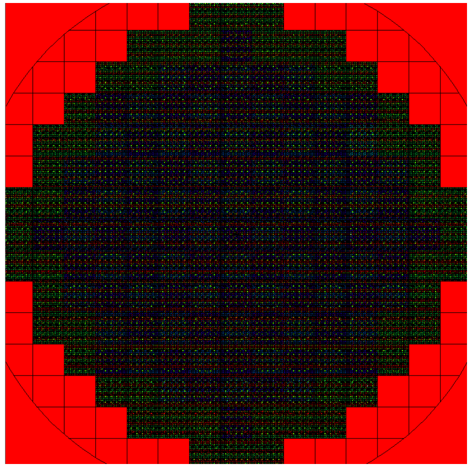
PWR Assembly



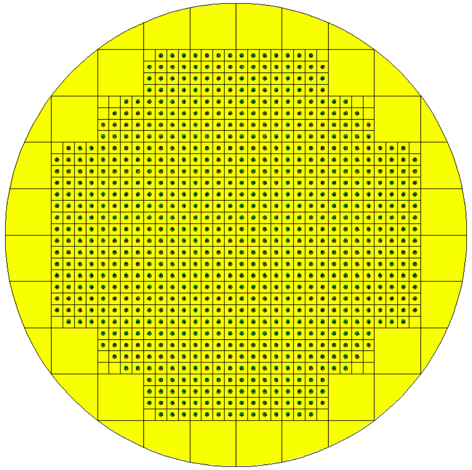
FBR



NRX

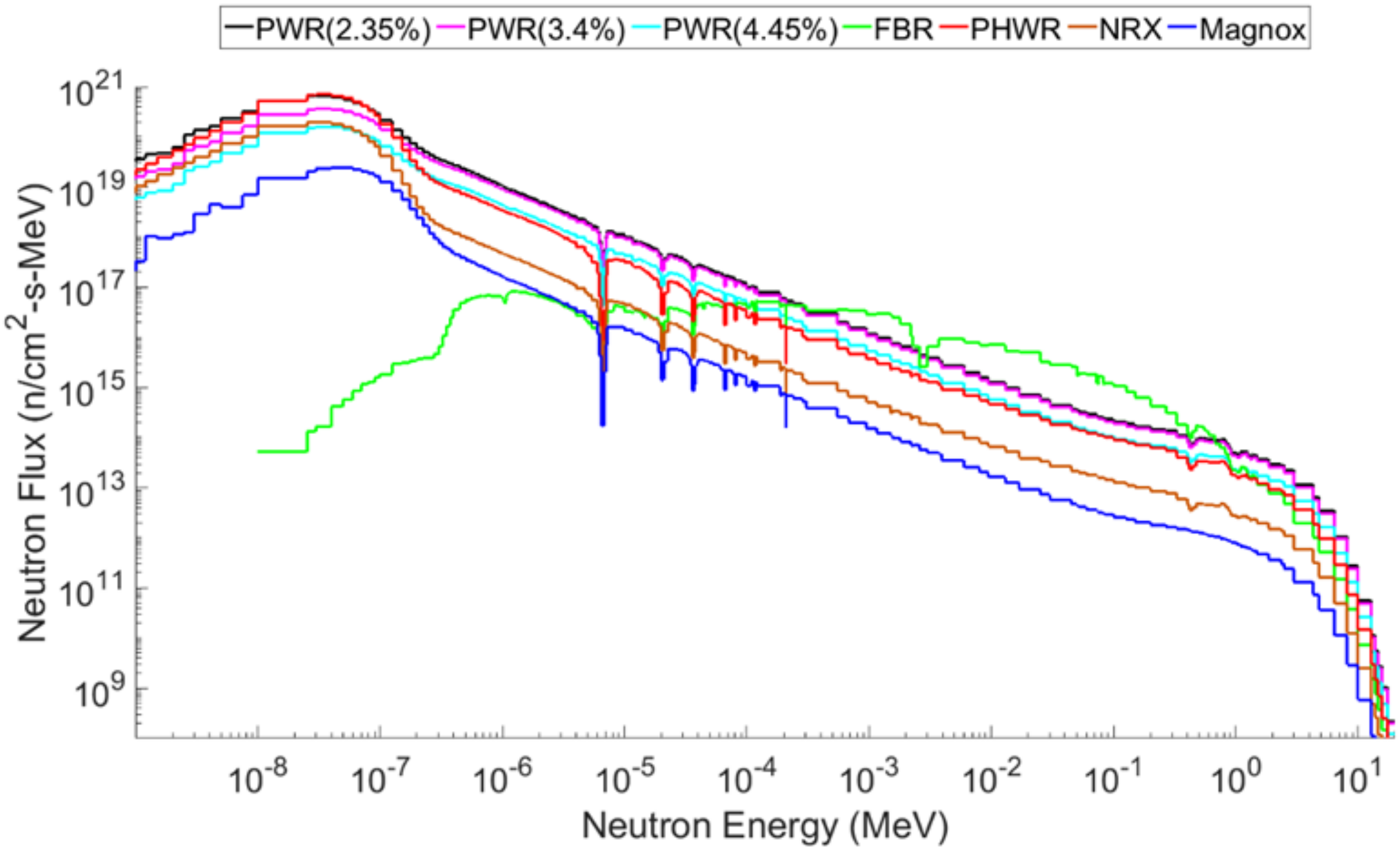


PWR



Magnox

Reactor Library Neutron Spectra



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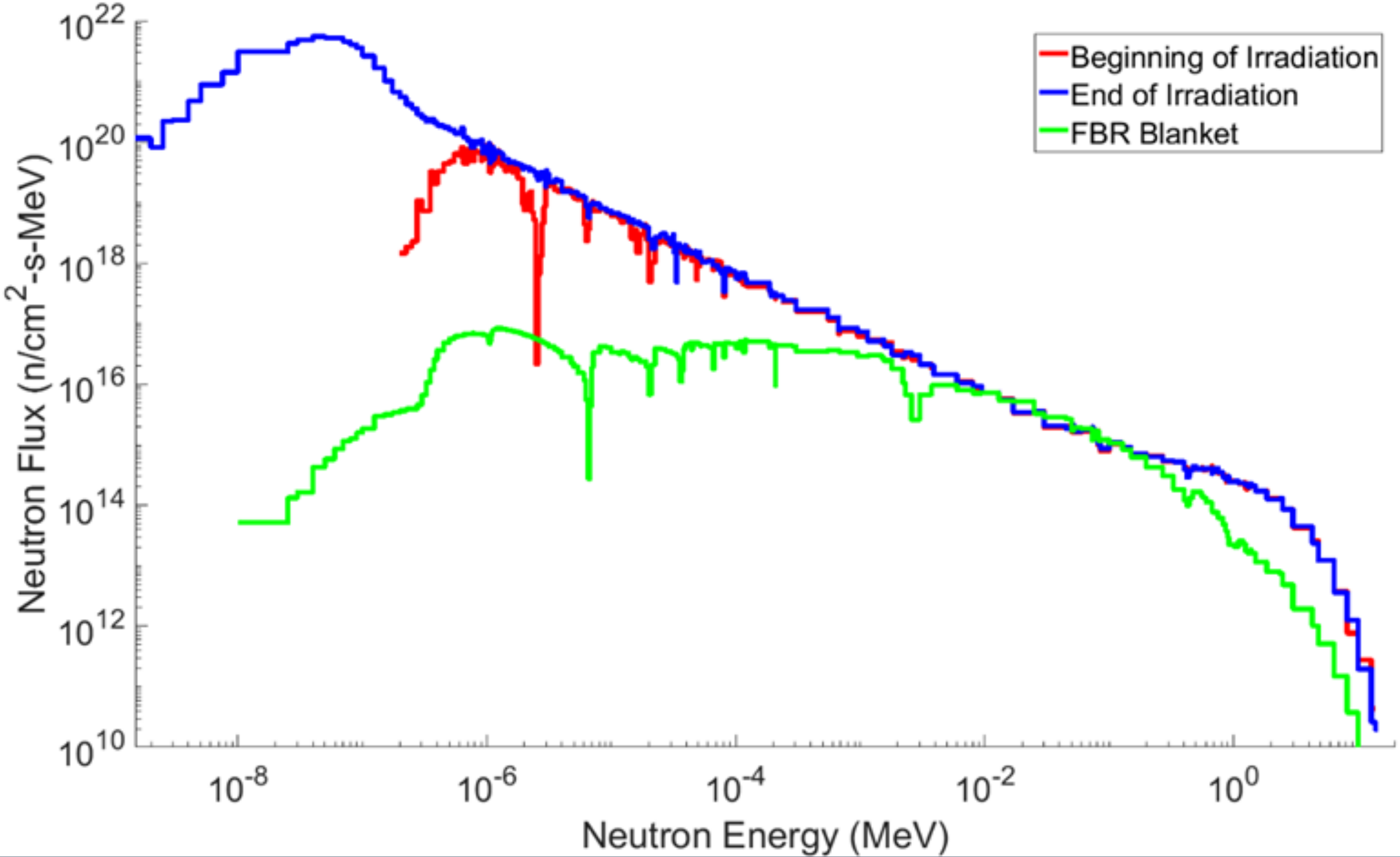
HFIR Irradiation Spectrum



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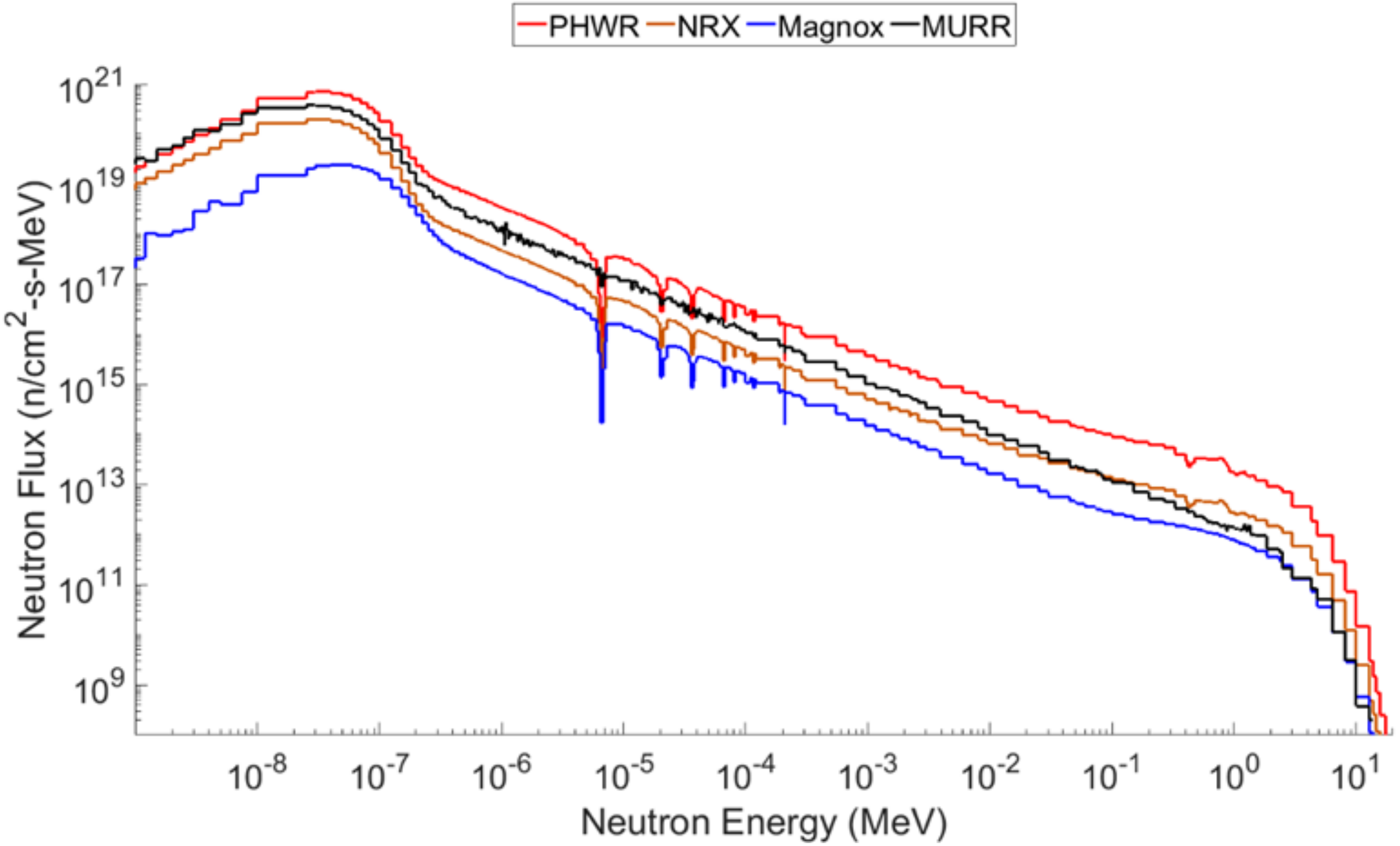
MURR Irradiation Spectrum



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MURR Irradiation Burnup

- Determination of burnup via ^{137}Cs measurements
- $A_n = \frac{CPS_n}{\varepsilon_\gamma * Y_\gamma}$

Fuel Disc	CPS (Live)	Measured ^{137}Cs Activity (Bq)	End of Irradiation ^{137}Cs Activity (Bq)
A	69.86	$1.65 \times 10^6 \pm 2.48 \times 10^4$	$1.67 \times 10^6 \pm 2.51 \times 10^4$
B	70.25	$1.66 \times 10^6 \pm 2.50 \times 10^4$	$1.68 \times 10^6 \pm 2.52 \times 10^4$
C	71.66	$1.70 \times 10^6 \pm 2.55 \times 10^4$	$1.71 \times 10^6 \pm 2.57 \times 10^4$

MURR Irradiation Burnup Cont.

- $Bu = \frac{N_{137Cs} * Q * GWd}{Y_{137} * U}$
  - Q = 202 ± 5 MeV
  - GWd = (1 GWd = 5.393 × 10^{26} MeV)
  - Y_{137} = 6.221% ± 0.069%
  - U = 14.52 mg ± 0.23 mg (1.452 × 10^{-9} MT)

Fuel Disc	Measured Burnup (GWd/MTU)	Measured Burnup Error	Simulated Burnup (GWd/MTU)	Simulated Burnup Error	S/E
A	0.949	3.32%	0.960	0.086%	1.01 ± 0.03
B	0.954	3.32%	0.960	0.086%	1.01 ± 0.03
C	0.973	3.32%	0.960	0.086%	0.99 ± 0.03

Irradiated Sample Dissolution Setup

Irradiated Sample Measurements

- Gamma spectrometry measurements using a Canberra HPGe
- Mass spectrometry measurements using a Thermo Fisher Scientific ICP-MS

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MURR Gamma Spectrometry

Isotope	Measured Activity in Full Disc (Bq)	Count Rate (CPS) Error	Simulated Activity in Full Disc (Bq)	S/E
^{95}Zr	2.54×10^7	< 0.1%	2.49×10^7	0.98
^{103}Ru	4.92×10^6	0.3%	5.62×10^6	1.14
^{134}Cs	1.12×10^5	2.2%	9.92×10^4	0.89
^{137}Cs	1.71×10^6	0.1%	1.67×10^6	0.98
^{141}Ce	4.21×10^6	0.1%	4.77×10^6	1.13
^{144}Ce	2.94×10^7	< 0.1%	3.11×10^7	1.06

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MURR Mass Spectrometry

Isotope	Fissiogenic Ratio	Measured Mass (g)	Measured Mass Relative Error	Simulated Mass (g)	S/E
^{133}Cs	1	5.22×10^{-7}	6.0%	5.42×10^{-7}	1.04
^{135}Cs	1	1.50×10^{-7}	6.2%	1.42×10^{-7}	0.94
^{137}Cs	0.976	5.08×10^{-7}	6.0%	5.14×10^{-7}	1.01
^{148}Nd	0.983	1.55×10^{-7}	5.8%	1.54×10^{-7}	0.99
^{149}Sm	1	8.34×10^{-9}	5.8%	9.51×10^{-9}	1.14
^{150}Sm	0.589	9.22×10^{-8}	5.8%	9.24×10^{-8}	1.00
^{152}Sm	1	5.55×10^{-8}	5.8%	5.58×10^{-8}	1.01
^{153}Eu	1	1.76×10^{-8}	5.9%	1.87×10^{-8}	1.06

MURR Mass Spectrometry - Plutonium

Measured Pu Mass (μg)	Measurement Error	Simulated Pu Mass (μg)	Simulated Stochastic Error	S/E
20.1	5.3%	20.9	0.78%	1.04 ± 0.06

Isotope	Measured Pu Vector	Measured Pu Vector Relative Error	Simulated Pu Vector	S/E[a]
^{239}Pu	95.22%	0.1%	95.75%	1.01
^{240}Pu	4.55%	2.2%	4.05%	0.89
^{241}Pu	0.23%	1.9%	0.20%	0.86
^{242}Pu	<0.01%	N/A	<0.01%	N/A

Maximum Likelihood Calculation

- Likelihood equation:

$$L(M, Bu, TSI|r_{mes}) \propto f(r_{mes}|M, Bu, TSI) = \prod_{j=1}^{n} \frac{1}{\sigma_{j,sim}\sqrt{2\pi}} exp\left\{-\frac{\left(r_{j,mes}-r_{j,sim}\right)^2}{2\sigma_{j,sim}^2}\right\}$$

- Log-likelihood equation:

$$Log\ L(M, Bu, TSI|r_{mes}) = \sum_{j=1}^{n}\left[log\left(\frac{1}{\sigma_{j,sim}\sqrt{2\pi}}\right) - \frac{\left(r_{j,mes}-r_{j,sim}\right)^2}{2\sigma_{j,sim}^2}\right]$$

- Variance in the log-likelihood:

$$\sigma_{Log\ L}^2 = \sum_{j=1}^{n}\left(\frac{\left(r_{j,mes}-r_{j,sim}\right)}{\sigma_{j,sim}^2}\right)^2 \times\left(\sigma_{j,mes}^2 + \sigma_{j,sim}^2\right)$$

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Maximum Likelihood Results – MURR

Results of the Maximum Likelihood Analysis for the MURR Irradiated Material [a]

Reactor Model	Log-Likelihood Value [b]	Predicted Burnup (GWd/MTU)	Predicted Time Since Irradiation (days)
MURR	+29.5 ± 1.1	1.02	295
NRX	+25.3 ± 3.0	1.03	208
MAGNOX	+13.2 ± 5.7	0.73	0
PWR (3.4%)	-6.02 ± 8.71	3.91	1381
PWR (4.45%)	-8.88 ± 10.2	≥ 3.90	1196
PWR (2.35%)	-12.7 ± 10.2	3.10	1202
PHWR	-14.7 ± 13.8	1.02	360
HFIR	-166 ± 28	4.40	1790
FBR	$-1.52 \times 10^5 \pm 2.02 \times 10^4$	≥ 4.73	0

[a] True Bu = 0.97 ± 0.03 GWd/MTU, True TSI = 318 days

[b] Maximum possible log-likelihood is 29.7

Task 3: Maximum Likelihood Results – HFIR

Results of the Maximum Likelihood Analysis for the HFIR Irradiated Material [a]

Reactor Model	Log-Likelihood Value [b]	Predicted Burnup (GWd/MTU)	Predicted Time Since Irradiation (days)
HFIR	+19.5 ± 4.6	4.29	1827
MURR	-46.6 ± 12.8	4.16	1700
NRX	-52.5 ± 12.5	4.13	1590
MAGNOX	-59.5 ± 13.3	3.00	421
PWR (2.35%)	-86.7 ± 21.0	≥ 5.31	1705
PHWR	-129 ± 32	≥ 4.35	2308
PWR (3.4%)	-284 ± 26	≥ 5.01	0
PWR (4.45%)	$-5.27 \times 10^3 \pm 1.38 \times 10^2$	≥ 3.90	0
FBR	$-6.39 \times 10^5 \pm 1.05 \times 10^4$	≥ 4.73	0

[a] True Bu = 4.36 ± 0.28 GWd/MTU, True TSI = 1601 days

[b] Maximum possible log-likelihood is 28.5

National Nuclear Security Administration

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